



(19) Europäisches Patentamt
 European Patent Office
 Office européen des brevets



(11) Publication number: 0 499 865 B1

(12)

EUROPEAN PATENT SPECIFICATION

(45) Date of publication of patent specification: 30.08.95 (51) Int. Cl. 6: C03C 14/00, C04B 35/58, C04B 35/80
 (21) Application number: 92101704.2
 (22) Date of filing: 03.02.92

(54) Glass-aluminum nitride composite material.

(30) Priority: 04.02.91 JP 33345/91

(43) Date of publication of application:
 26.08.92 Bulletin 92/35

(45) Publication of the grant of the patent:
 30.08.95 Bulletin 95/35

(54) Designated Contracting States:
 DE FR GB IT

(56) References cited:
 EP-A- 0 374 825

PATENT ABSTRACTS OF JAPAN vol. 14, no. 531 (C-780)21 November 1990 & JP-A-2 221 162

WORLD PATENTS INDEX LATEST Week 8909, Derwent Publications Ltd., London, GB; AN 89-065572 & JP-A-1 018 102

CHEMICAL ABSTRACTS, vol. 113, no. 10, 3 September 1990, Columbus, Ohio, US; abstract no. 83768G, page 337 & JP-A-2 107 571

(73) Proprietor: SUMITOMO ELECTRIC INDUSTRIES, LTD.
 5-33, Kitahama 4-chome,
 Chuo-ku
 Osaka-shi,
 Osaka 541 (JP)

(72) Inventor: Yamakawa, Akira, c/o Itami Works of Sumitomo Elec. Industries, Ltd,
 1-1, Koyakita 1-chome
 Itami-shi,
 Hyogo (JP)
 Inventor: Sogabe, Koichi, c/o Itami Works of Sumitomo Elec. Industries, Ltd,
 1-1, Koyakita 1-chome
 Itami-shi,
 Hyogo (JP)

(74) Representative: Flener, Josef
 Patentanwälte Kahler, Käck, Flener & Sturm
 P.O. Box 12 49
 D-87712 Mindelheim (DE)

Note: Within nine months from the publication of the mention of the grant of the European patent, any person may give notice to the European Patent Office of opposition to the European patent granted. Notice of opposition shall be filed in a written reasoned statement. It shall not be deemed to have been filed until the opposition fee has been paid (Art. 99(1) European patent convention).

CHEMICAL ABSTRACTS, vol. 110, no. 6, 6
February 1989, Columbus, Ohio, US; abstract
no. 43704Q, page 300 & JP-A-63 210 043

CHEMICAL ABSTRACTS, vol. 112, no. 16, 16
April 1990, Columbus, Ohio, US; abstract no.
144519E, page 323 & JP-A-1 219 066

CHEMICAL ABSTRACTS, vol. 112, no. 16, 16
April 1990, Columbus, Ohio, US; abstract no.
144489V, page 322 & JP-A-1 179 740

PATENT ABSTRACTS OF JAPAN vol. 12, no.
501 (C-556)27 December 1988 & JP-A-63 210
043

Description**BACKGROUND OF THE INVENTION**5 **1. Field of the Invention**

The present invention relates to a glass-aluminum nitride composite having a high heat conductivity, particularly a glass-aluminum nitride composite having properties suitable for a semiconductor packaging material.

10

2. Description of the Prior Art

An increase in the speed of electronics elements and an increase in the integration density have been rapidly advanced. This has led to a severer demand for an insulating substrate on which the elements are mounted and packaging material. A high heat conductivity for coping with an increase in the density of heat buildup, a low dielectric constant for coping with an increase in the speed, and a lowering in the conductor resistance are particularly required of the packaging material. Regarding the highly heat conductive material, development has been made on an AlN material in addition to BeO and SiC. On the other hand, the demand for a lowering in the dielectric constant and a lowering in the conductor resistance cannot be satisfied by the highly heat conductive material, and priority has been given to the development of a glass-ceramic composite material. In the glass-ceramic composite material, both the heat conductivity and the mechanical strengths are so low that no satisfactory properties are obtained. For this reason, in order to meet the demand, studies have been made on a composite material comprising glass and a highly heat conductive powder. For example, Japanese Patent Laid-Open No. 210043/1988 disclosed a composite comprising glass and aluminum nitride powder. This composite has a permittivity of 5 to 7 and a heat conductivity of 20 W/m·K. Further, in Japanese Patent Laid-Open Nos. 221162/1990 and 196073/1990, corresponding to EP-A-0 374 825, a further slight improvement in the heat conductivity could be attained by specifying the oxygen content in aluminum nitride.

The above-described conventional glass-aluminum nitride composite is still unsatisfactory not only in the improvement in the heat conductivity but also in the strength of the substrate, and the improvement achieved therewith is at the most that of the performance of the conventional low temperature fired substrate, so that this composite cannot satisfy a further advanced property requirement. In the present invention, the heat conductivity is further improved particularly in a glass composite having a low permittivity and a high heat conductivity and provides a material having a high strength besides the above-described properties.

SUMMARY OF THE INVENTION

The present invention relates to a glass-aluminum nitride composite material comprising a sintered body produced by adding glass powder to aluminum nitride grains having an oxygen content of less than 2% by weight and a mean grain diameter of 1.0 μm or more and subjecting the mixture to molding and sintering, the glass-aluminum nitride composite material having a heat conductivity of 30 W/m·K or more and the content of AlN is 50 to 95% by weight based on the whole composite material. The glass powder preferably comprises 30 to 60 wt.% of SiO₂, 10 to 30 wt.% of Al₂O₃, 10 to 30 wt.% of B₂O₃ and 30 wt.% of MO (wherein M represents Mg, Ca or Sr). The addition of an amount of AlN whiskers thereto contributes to a further improvement in the strength.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

50 The AlN grain used in the present invention is required to have an oxygen content of less than 2% by weight. When the oxygen content is 2 wt.% more, the heat conductivity of the composite is so insufficient that effect attained by compositing AlN becomes small. The mean grain diameter is 1.0 μm or more. When it is less than 1.0 μm , the lowering in the heat conductivity is significant due to the diffusion of an element from the matrix glass, so that it is necessary to use a raw material having an at least given grain diameter.

55 The lower the oxygen content, the better the results. In particular, a good heat conductivity can be obtained when the oxygen content is 0.4% by weight or less. When the grain diameter exceeds 10.0 μm , no surface accuracy after sintering can be ensured. Further, it is possible to incorporate an amount of AlN whiskers for the purpose of increasing the strength of the composite. For example, it is useful to add an amount of AlN

whiskers having a minor diameter of 0.3 μm and a length of about 5 μm in an amount of 5% by volume. The glass powder having the above-described composition constituting the glass matrix is the powder of a known borosilicate glass which is preferred from the viewpoint of sintering temperature, permittivity, etc. The composition is regulated in the above-described range to regulate the sintering temperature. Further, 5 use may be made of a paste having a low electric resistance, such as Ag, Au or Cu, and simultaneous firing can be conducted to form a conductive circuit. Further, it is also possible to regulate the permittivity.

Example 1:

10 Each of the AlN samples having properties specified in Table 1 and glass powder (having a composition consisting of 50 wt.% of SiO_2 , 15 wt.% of Al_2O_3 , 20 wt.% of B_2O_3 and 15 wt.% of CaO) were mixed with an organic binder, etc., in a ball mill according to the formulation specified in the Table to 15 prepare a slurry. The slurry was molded into a tape and fired in a nitrogen stream at 900 $^\circ\text{C}$. The resultant glass-aluminum nitride composite had a high heat conductivity as given in Table 2. It was possible to provide internal wiring and lamination before the firing. Further, it was also possible to bake a paste of metal, such as Au, Ag or Cu, in the surface of the sintered body. These render the composite useful as an electrical wiring board.

Table 1

20

No.	Properties of AlN		AlN content (wt.%)
	oxygen content (wt.%)	mean grain diameter (μm)	
25	1	0.3	65
	2	0.8	65
	3	1.5	65
	4*	2.5	65
	5*	1.5	65
30	6	1.5	65
	7	1.5	80
	8	1.5	60
	9*	1.5	40
35	10	0.3	60
	11	0.4	60
	12	0.5	60
	13	0.8	60
	14*	AlN sintered body	

40

Note) *: comparative example

45

50

55

Table 2

No.	Volume specific resistance $\Omega \cdot \text{cm}$	Heat conductivity $\text{W/m} \cdot \text{K}$	Relative permittivity	Bending strength kg/mm^2
1	$>10^{13}$	35	5.0	25
2	do.	45	do.	do.
3	do.	40	do.	do.
4*	do.	10	do.	do.
5*	do.	12	do.	20
6	do.	45	do.	25
7	do.	50	5.5	do.
8	do.	35	5.0	do.
9*	do.	15	4.5	20
10	do.	50	5.0	25
11	do.	do.	do.	do.
12	do.	45	do.	do.
13	do.	40	do.	do.
14*	do.	180	8.0	35

20 Note) *: comparative example

Example 2:

25 A sintered body was prepared in the same manner as that of sample No. 3 of the Example 1, except that 5% by weight, based on the total weight of the glass and the ceramic component, of an amount of AlN whiskers (major diameter: 10 μm , minor diameter: 0.4 μm , transparent) was further added to the slurry.

30 The resultant sintered body had a bending strength of 40 kg/mm^2 , and no deterioration was observed in the volume specific resistance, heat conductivity and relative permittivity.

According to the present invention, it is possible to provide a material having a high heat conductivity, a low permittivity and a high strength. This material is suitable for use in a semiconductor packaging material.

Claims

1. A glass-aluminum nitride composite material comprising a sintered body produced by adding glass powder to aluminum nitride grains having an oxygen content of less than 2% by weight and a mean grain diameter of 1.0 μm or more, mixing the glass powder and the AlN grains preferably with a binder, and subjecting the mixture to molding and sintering, said glass-aluminum nitride composite having a heat conductivity of 30 $\text{W/m} \cdot \text{K}$ or more and the content of AlN is 50 to 95% by weight based on the whole composite material.
2. A glass-aluminum nitride composite material according to claim 1, wherein the glass powder consisting of 30 to 60% (% by weight; the same shall apply hereinafter) of SiO_2 , 10 to 30% of Al_2O_3 , 10 to 30% of B_2O_3 and 30% or less of MO (wherein M represents Mg, Ca or Sr)
3. A glass-aluminum nitride composite material according to claims 1 or 2, whereto an amount of AlN whiskers is further added.
4. A glass-aluminum nitride composite material according to any of claims 1 to 3, wherein the oxygen content of the AlN grains is less than 0.5 % by weight.
5. A glass-aluminum nitride composite material according to any of claims 1 to 4, wherein the grain diameter is 1.0 to 10.0 μm .
6. A glass-aluminum nitride composite material according to any of claims 3 to 5, wherein the AlN whiskers have a diameter of about 0.3 μm and a length of about 5 μm and are added in an amount of 5% by volume.

Patentansprüche

1. Ein Glas-Aluminiumnitrid-Verbundwerkstoff aufweisend einen Sinterkörper, der hergestellt wird durch Zugabe von Glaspulver zu Aluminiumnitridkörnern, die einen Sauerstoffgehalt von weniger als 2 Gewichts-% und einen durchschnittlichen Korndurchmesser von 1,0 μm oder mehr aufweisen, Mischen des Glaspulvers und der AlN-Körner, vorzugsweise mit einem Bindemittel, und Unterziehen der Mischung einer Formgebung und Sinterung, wobei die Glas-Aluminiumnitrid-Zusammensetzung eine Wärmeleitfähigkeit von 30 W/m•K oder mehr aufweist und der AlN-Gehalt 50 bis 95 Gewichts-% beträgt, basierend auf dem gesamten Verbundwerkstoff.
2. Ein Glas-Aluminiumnitrid-Verbundwerkstoff nach Anspruch 1, wobei das Glaspulver 30 bis 60% (Gewichts-%; gleiches gilt im nachfolgenden) an SiO_2 , 10 bis 30% an Al_2O_3 , 10 bis 30% an B_2O_3 und 30% oder weniger an MO aufweist, (wobei M Mg, Ca oder Sr darstellt).
3. Ein Glas-Aluminiumnitrid-Verbundwerkstoff nach Anspruch 1 oder 2, wobei zusätzlich eine Teilmenge an AlN-Whiskern zugegeben ist.
4. Ein Glas-Aluminiumnitrid-Verbundwerkstoff nach einem der Ansprüche 1 bis 3, wobei der Sauerstoffgehalt der AlN-Körner kleiner als 0,5 Gewichts-% ist.
5. Ein Glas-Aluminiumnitrid-Verbundwerkstoff nach einem der Ansprüche 1 bis 4, wobei der Korndurchmesser 1,0 bis 10,0 μm beträgt.
6. Ein Glas-Aluminiumnitrid-Verbundwerkstoff nach einem der Ansprüche 3 bis 5, wobei die AlN-Whisker einen Durchmesser von ca. 0,3 μm und eine Länge von ca. 5 μm aufweisen und in einer Teilmenge von 5 Volumen-% zugegeben sind.

Revendications

1. Matériau composite en verre-nitrite d'aluminium comprenant un corps fritté produit en ajoutant de la poudre de verre à des grains de nitrite d'aluminium ayant une teneur en oxygène inférieure à 2% en poids et une granulométrie de 1,0 μm ou plus, en mélangeant la poudre de verre et les grains de AlN de préférence en présence d'un liant et en soumettant le mélange à un moulage et à un frittage, ledit composite en verre-nitrite d'aluminium ayant une conductivité thermique de 30 W/m.K ou plus et la teneur en AlN étant de 50 à 95% en poids par rapport à la quantité globale de matériau composite.
2. Matériau composite en verre-nitrite d'aluminium selon la revendication 1, dans lequel la poudre de verre est constitué de 30 à 60% (en poids; il en est de même ci-après) de SiO_2 , de 10 à 30% de Al_2O_3 , de 10 à 30 % de B_2O_3 et de 30 % ou moins de MO (dans lequel M représente Mg, Ca ou Sr).
3. Matériau composite en verre-nitrite d'aluminium selon la revendication 1 ou 2, auquel une certaine quantité de fibres de renfort en AlN est également ajoutée.
4. Matériau composite en verre-nitrite d'aluminium selon l'une quelconque des revendications 1 à 3, dans lequel la teneur en oxygène des grains de AlN est inférieure à 0,5% en poids.
5. Matériau composite en verre-nitrite d'aluminium selon l'une quelconque des revendications 1 à 4, dans lequel le diamètre des grains est de 1,0 à 10,0 μm .
6. Matériau composite en verre-nitrite d'aluminium selon l'une quelconque des revendications 3 à 5, dans lequel les fibres de renfort en AlN présentent un diamètre d'environ 0,3 μm et une longueur d'environ 5 μm et sont ajoutées en quantité de 5% en volume.